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Stoichiometry and atomic concentration depth profiles in InAs/Si quantum dot systems by Rutherford backscattering spectroscopy and secondary ion mass spectroscopy

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Abstract. Stoichiometry and atomic concentration distributions for MBE grown systems of InAs quantum dots buried in a Si matrix has been studied using Rutherford Backscattering spectroscopy and Secondary Ion Mass Spectroscopy depth profiling. We have found a significant excess of As atoms which under elevated temperatures diffuse into the Si matrix despite of a larger ionic radius. This may affect the band alignment on the QD/matrix interface. Our findings suggest improvements of the MBE growth process for InAs/Si and similar systems.

Si based devices predominate modern microelectronics. This material can however not be immediately used for light-emitting devices due to its indirect band gap, which limits the luminescence efficiency. Several approaches are under development to incorporate optoelectronics into the widespread Si technology. One of them is to embed semiconductor nanocrystals having a narrow direct band gap into a Si matrix. Accumulation of the electrons and holes in the nanocrystals requires a proper band alignment relative to the matrix, which in many cases is critically influenced by a strain imposed by a lattice mismatch [1]. It is anticipated, that the luminescence efficiency can be increased, if an atomlike discrete electronic spectrum is formed by the confining potential in the nanocrystals - quantum dots (QDs).

Characterization of QD nanocrystals is a challenging task, because they are normally buried under a capping layer having a thickness of some tens of nanometers. Usually Transmission Electron Microscopy (TEM) is used, but it is less sensitive to the stoichiometry and atomic concentration distributions, including point defects (e.g. interstitial inclusions). Here we report on studies of these aspects of InAs/Si QD systems using Rutherford Backscattering Spectroscopy (RBS) and Secondary Ion Mass Spectroscopy (SIMS). The first method is able to reach buried atomic layers non-destructively and to quantitatively determine their atomic content.

We report on RBS and SIMS measurements on a system of InAs nanocrystals grown by MBE in a Si matrix [1]. This system is particularly interesting, because a strain induced by the lattice mismatch between InAs and Si is presumably crucial for formation of the band alignment required for the carrier accumulation in the QDs. The obtained results on stoichiometry and atomic concentrations are helpful for further optimization of the growth process for these and similar systems.

The InAs/Si samples were grown on highly-oriented Si(100) substrates using an EP1203 (Russia) MBE machine. Thermal desorption of native silicon oxide before growth was achieved by heating the substrate up to 830°C-870°C for 15 min. After that well resolved

(2×1) or mixed (1×2) and (2×1) surface reconstructions, typical for cleaved Si(100) surfaces, were observed. The substrate was then cooled down to the required growth temperature, and deposition of InAs was initiated. The InAs deposition rate was typically 0.1 monolayers (ML) per second. Calibration of the growth rate, the As-to-In flux ratio, and monitoring of the surface morphology during growth was performed using Reflection High-Energy Electron Diffraction (RHEED). The as-grown InAs nanocrystals, as revealed by STM [2], are pyramids with lateral dimensions of $\sim 12 \times 20$ nm and a heights of ~ 4 nm. They are placed randomly and occupy $\sim 50\%$ of the substrate area. Just after the InAs deposition, a 10–20 nm thick Si capping layer was grown at the same substrate temperature. The growth was followed by annealing at 650–700°C lasting 10 min. Then an additional 20–40 nm thick Si capping layer was grown at the same temperature. It was followed by annealing at 700–800°C for 10 min in order to activate migration of the surface atoms and smoothen the resulting Si surface. The grown samples were characterized by photoluminescence (PL) spectroscopy, which revealed spectral peaks originated from the InAs QDs [3].

The RBS measurements were performed on a few InAs/Si samples with a nominal InAs layer thickness of 2 and 2.5 monolayers, respectively, and a Si capping layer thickness of ~ 15 nm. He^+ ions with the energy of 1.5 MeV were used in the primary beam, which allowed to probe to depths much larger than the thickness of the capping layer. A typical RBS spectrum is shown in Fig. 1. It is taken for a representative sample containing a 2.5 ML thick InAs layer grown at 400°C, which was capped by a 16 nm thick Si layer grown at approximately the same temperature and annealed at 780°C for 10 min. The spectrum reveals the depth distribution and the amount of material of the different atomic species. The measured spectra shows well-separated peaks of As and In (see the main pane of Fig. 1) and a steep increase in the backscattering yield at lower energies due to the Si matrix (insert). The ratio of the areas below the As and In peaks, weighted with the corresponding backscattering cross-sections proportional to Z^2 , directly gives the ratio of the number of the As to In atoms incorporated.

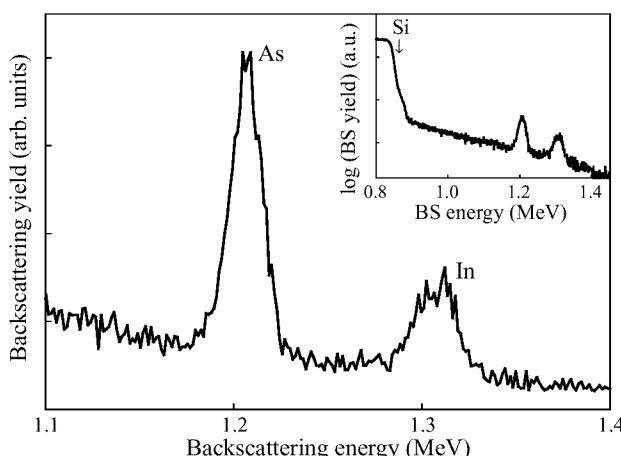


Fig. 1. A portion of the RBS spectrum taken on a representative InAs/Si sample, showing the signals of As and In. The insert shows the entire energy range. The steep increase in backscattering yield below 0.9 MeV is due to the Si matrix.

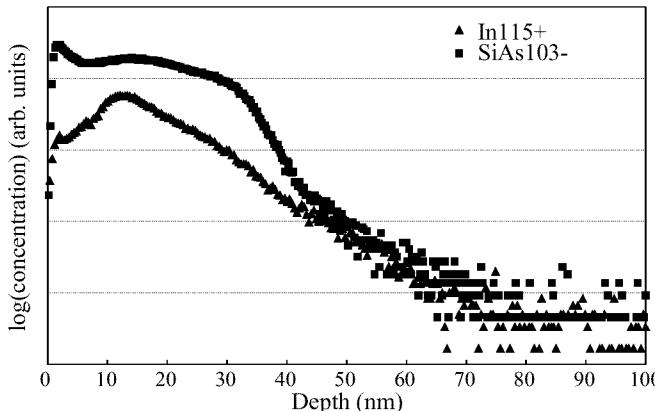


Fig. 2. SIMS depth profiles of $^{115}\text{In}^+$ measured by O^{2+} sputtering and $^{103}\text{SiAs}^+$ measured by Cs^+ sputtering. In both cases the ion beam energy was 3 keV. The sample is the same as in Fig. 1. A clear change in the slope of the As profile indicates diffusion of excess As atoms into the Si substrate.

The In-to-As ratio $N_{\text{As}}/N_{\text{In}}$ was determined within the accuracy of about 1%. Surprisingly, we have found that there was ~ 5 times more As than In and even more, depending on the preparation condition. As the nanocrystalline InAs in the QDs have nevertheless the ideal stoichiometry, as indicated by high-resolution TEM [4], the excess As atoms must be diluted in the surrounding epitaxially grown Si capping layer and substrate without affecting the properties of the QDs. Apart from possible small excess of As during growth of the InAs layer, these As atoms were apparently built in during MBE growth of the capping layers, as the partial pressure of As_4 after closing the shutter of the source decreases rather slowly. Due to elevated temperatures during the growth and annealing steps the As atoms could then diffuse over the capping layer and even into the substrate.

The elemental depth distributions could also be extracted from the RBS spectra using a fitting procedure. We however used for this purpose dynamic SIMS depth profiling which has a higher depth resolution. The area density of As and In, evaluated from a fit to the RBS-spectra, was used to calibrate the SIMS depth profiles in absolute In and As concentrations allowing thus their direct comparison. The depth scale was calibrated using stylus profiler measurements of the sputtered crater depths. Typical SIMS-spectra of In and As are shown in Fig. 2.

The SIMS-profiles within a depth region of ~ 5 nm from the surface are distorted by surface effects due to non-stationary sputtering conditions and a layer of native silicon oxide formed during storage in air. Upon passing of the InAs layer the In and SiAs signals do not drop abruptly, because a certain amount of sputtered material remains on the surface and is driven into the surface by knock-on collisions. However, a distinct peculiarity of the As spectrum is that it decays to larger depths with two different rates. While the background decay can be explained by the knock-on collisions, the slower decay next to the maximum indicates diffusion of As atoms into the substrate. This finding is particularly interesting in view of the As atomic radius being larger than the Si one. It hints to an anomalous diffusion mechanism of As in Si. This effect, previously neglected, should be taken into account in the MBE growth. For example, a pause after the InAs growth followed by flashing off the adsorbed As atoms could be introduced before deposition of the Si capping layer. The excess As atoms can act as additional dopants in the Si matrix around the QDs and thus

influence the band alignment.

Summarizing, we have determined the stoichiometry and atomic concentration distributions in the MBE grown samples of InAs QDs buried in a Si matrix using non-destructive RBS measurements and SIMS depth profiling. A significant excess of As atoms built in during the growth of the capping layers has been found. Under elevated growth and annealing temperatures the As atoms diffuse into the Si matrix despite of their larger ionic radius. These findings can help to optimize the MBE growth process for InAs/Si and similar systems.

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